

# Tunnel effect in ferromagnetic half-metal Co<sub>2</sub>CrAl-superconductor heterostructures

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Ferromagnetic half-metal Co<sub>2</sub>CrAl films and tunnel contacts Co<sub>2</sub>CrAl - insulator (I) - Pb are fabricated and investigated. It is found that the normalized differential conductivity  $\sigma^{\text{FS}}$  of such tunnel junctions with low resistance is larger than the normalized differential conductivity  $\sigma^{\text{NS}}$  of known normal metal - I - superconductor type tunnel junctions. It is shown that the observed increase in  $\sigma^{\text{FS}}$  is caused by the accumulation of spin polarized electrons in a superconductor and can be used for estimating the spin polarization degree  $P$  in ferromagnets. This method shows that  $P$  of L2<sub>1</sub>-type ordered Co<sub>2</sub>CrAl Heusler alloy films at  $T = 4.2$  K is close to 1.

*Key words:* spin polarization, spin current, effective chemical potential, differential conductivity, nonequilibrium superconductivity.

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## I. INTRODUCTION

During the recent years spintronics has become a rapidly developing science. Therefore, studying the peculiarities of spin-polarized current injection into superconductors (S) is an actual task.<sup>1-3</sup>

Among possible candidates for spin-polarized current injectors from a ferromagnet (F) into a superconductor some of Heusler alloys (HA) seem to be more preferable. Indeed, some of the full HA of X<sub>2</sub>YZ composition (here X and Y are 3d transition metals, and Z is *s-p* metal) are ferromagnets and show significant, up to 100%, spin polarization  $P$  due to the deep minimum in the energy band gap for the minority spin electrons at the Fermi level ( $E_F$ ).<sup>4-6</sup> In particular, some of the Co-based HA (and Co<sub>2</sub>CrAl is among them) are half-metals with high Curie temperature and high magnetic moment.<sup>7-9</sup> Polycrystalline Co<sub>2</sub>CrAl alloy ingots have been fabricated and investigated.<sup>10</sup> However, the measured spin polarisation  $P$  was 62% that was much less than the theoretical value. It is also necessary to mention that for the spin injectors, except the high degree of polarisation, their fabrication in the form of thin films is very important. Recently there has been a growing interest in investigating the F-S hybrid structures. They allow us to receive information on the ferromagnet spin polarisation, understand the influence of the spin polarized current on a superconducting state, and establish the physical ground for development of the spin criolectronics.

In this work we have fabricated quasimonocrystalline Co<sub>2</sub>CrAl films and the tunnel junctions of half-metal ferromagnet Co<sub>2</sub>CrAl (HMF)/insulator (I) /Pb (S), *i.e.* HMF/I/S structures and investigated the effect of injecting the spin-polarized electron current (SPE-current) into superconductor on its superconducting state.

## II. EXPERIMENT

The tunnel HMF/I/S junctions have a cross-like shape and  $200 \times 200 \mu\text{m}^2$  junction area. The spin-injectors, L2<sub>1</sub> - type ordered Co<sub>2</sub>CrAl alloy layers of about 100 nm in thickness, was deposited first by flash evaporation onto sapphire substrates kept at different temperatures in a vacuum better than  $2 \times 10^{-5}$  Pa. An insulating barrier layer on the top of the Co<sub>2</sub>CrAl films was formed by the natural oxidation of Co<sub>2</sub>CrAl layers at the ambient conditions. On the top of the insulating layer a Pb film of 100 - 200 nm in thickness was thermally deposited. The microscopic structures of Co<sub>2</sub>CrAl and Pb films were investigated by selective-area microdiffraction of transmission electron microscopy (TEM). It was shown that Co<sub>2</sub>CrAl and Pb films have L2<sub>1</sub>- type of structure and *fcc*-type of lattice, respectively (see Fig. 1). The magnetic properties of such prepared Co<sub>2</sub>CrAl films were investigated in a temperature range of 5 - 350 K using SQUID-magnetometer. Additionally, in-plane magnetic field dependences of magnetization  $M(H)$  were obtained using the vibrating sample magnetometer. Temperature dependence of magnetization for Co<sub>2</sub>CrAl obtained for cooled and measured at 100 Oe magnetic field (see Fig. 2) reveals the Curie temperature 330 K, *i.e.* close to that of the bulk sample.<sup>9</sup>

Thus fabricated Pb films exhibit the transition into the superconducting state at  $T = 7.2$  K with the critical current density  $j_c^{\text{Pb}}(4.2 \text{ K}) = 3 \times 10^6 \text{ A/cm}^2$ . Pb as superconductor was chosen for the following reasons. (1) Pb is the well investigated superconductor with *s*-type symmetry of the order parameter. (2) It has rather high critical temperature (7.2 K) and this fact allows us to obtain high resolution of superconducting parameters at liquid helium temperatures. (3) Diffusion of Pb into the barrier and the HMF layer is inhibited because of its rather large ion-radii. (4) Pb is chemically inactive.<sup>11</sup>

We have found that for these Co<sub>2</sub>CrAl-I-Pb junctions the value of normalized conductivity  $\sigma^{\text{FS}} \equiv G^{\text{FS}}/G^{\text{FN}}$  essentially differs from the value of fundamental normalized Giaever conductivity  $\sigma^{\text{NS}} \equiv G^{\text{NS}}/G^{\text{NN}}$  for tunnel junction

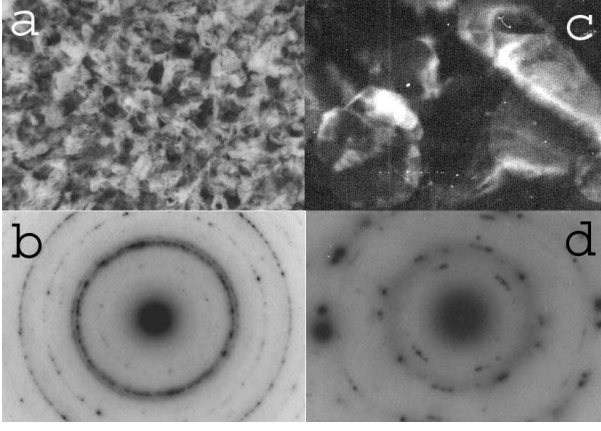


FIG. 1: The structure (a,c) and selective-area microdiffraction (b,d) of TEM for  $\text{Co}_2\text{CrAl}$  (a,b) and Pb (c,d) films.

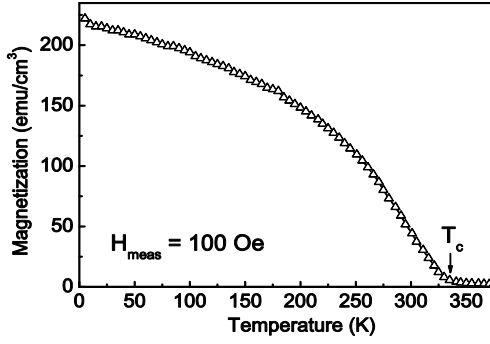


FIG. 2: Temperature dependence of magnetization obtained in FC regime for  $\text{Co}_2\text{CrAl}$  films.

of N-I-S type,<sup>12–14</sup> either calculated within the framework of Bardeen-Cooper-Schrieffer (BCS) theory or determined in experiment. (Here N is normal metal,  $G^{\text{FS}}$  and  $G^{\text{NS}}$  - differential conductivities of the tunnel junction at zero bias,  $G^{\text{FN}}$  and  $G^{\text{NN}}$  - differential conductivities of the same tunnel junction with S in the normal state). Besides, we have found that the value of  $\sigma^{\text{FS}}$  depends on the value of conductivity  $G^{\text{FN}}$ , while the value of  $\sigma^{\text{NS}}$  does not depend on  $G^{\text{NN}}$ .<sup>11–13</sup>

Fig. 3 shows the experimental results for normalized conductivity  $\sigma^{\text{FS}}$  of  $\text{Co}_2\text{CrAl}$ -I-Pb tunnel junctions investigated at temperature 4.2 K, which shows the aforementioned effects. One can see that as  $(G^{\text{FN}})^{-1}$  increases, the experimental value of  $(\sigma^{\text{FS}})^{-1}$  changes from 6 up to 100. Calculation within the framework of BCS theory for N-I-Pb junction at temperature 4.2 K gives  $(\sigma^{\text{NS}})^{-1} \approx 6.5$ . For the N-I-S type of Sn-I-Pb junction it was experimentally observed that  $(\sigma^{\text{NS}})^{-1} \approx 5.9$ ,<sup>15,16</sup> and for Al-I-Pb junction -  $(\sigma^{\text{NS}})^{-1} \approx 5.8$ .<sup>13</sup>

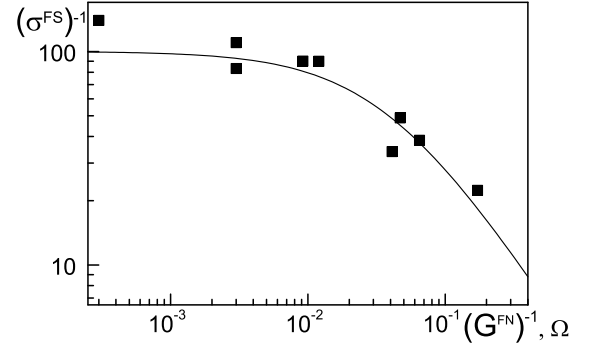


FIG. 3: Experimental data (squares) for normalized conductivity  $(\sigma^{\text{FS}})^{-1}$  dependence on conductivity  $(G^{\text{FN}})^{-1}$  for a set of  $\text{Co}_2\text{CrAl}$ -I-Pb tunnel junctions in the normal state at  $T = 4.2$  K, and theoretical dependence (solid line) for  $P = 0.97$ ,  $\Theta \sim (2G^{\text{FN}})^{-1}$ .

### III. DISCUSSION

For N-I-S tunnel junctions dependence of the tunnel current  $I$  on bias  $V$  at values  $eV$  smaller than the height of the potential barrier is determined as follows:<sup>14</sup>

$$I^{\text{NS}}(V) = C \int_{-\infty}^{+\infty} N_t(E_k) [f(E_k) - f(E_k + eV)] dE_k, \quad (1)$$

where  $C = e^{-1} G_{\text{NN}}$ ,  $G_{\text{NN}} = \frac{4\pi e^2}{\hbar} |T|^2 N^{(1)}(0) N^{(2)}(0)$  is conductance of the junction with both electrodes in the normal state,  $T$  is the tunneling matrix element,  $N^{(1)}(0)$ ,  $N^{(2)}(0)$  - densities of electronic states at the Fermi level in the junction banks,  $e$  - the elementary charge,  $N_t(E_k) = \text{Re} \frac{|E_k|}{\varepsilon_k}$  - quasiparticles density of states in the superconductor according to BCS theory,  $f(E_k)$  - Fermi-function of electronic states distribution with a momentum  $k$  and energy  $\varepsilon_k = \sqrt{E_k^2 - \Delta^2}$  for a superconductor with the energy gap  $\Delta$ .

At  $eV \ll \Delta$  the normalized differential conductivity  $\sigma^{\text{NS}}$  of N-I-S tunnel junctions can be calculated as<sup>14</sup>

$$\sigma^{\text{NS}}(T) = \frac{(dI/dV)_S}{(dI/dV)_N} = \int_{-\infty}^{+\infty} N_t(E) \frac{-\partial f(E)}{\partial E} dE. \quad (2)$$

If we replace the normal metal N in an N-I-S junction with a ferromagnet F, the dependence (1) of the current in an F-I-S tunnel junction will become as follows:

$$I^{\text{FS}}(V) = \sum_{\sigma} C_{\sigma} \int_{-\infty}^{+\infty} N_t(E_k) [f(E_k) - f(E_k + eV)] dE_k, \quad (3)$$

where  $eC_{\sigma} = \frac{2\pi e^2}{\hbar} |T|^2 N_{\sigma}^{(1)}(0) N^{(2)}(0)$  - junction conductivity for separate spin subzone of ferromagnet for normal

state of both electrodes; spin index  $\sigma$  runs over the values  $+1(\uparrow)$  and  $-1(\downarrow)$ ;  $N_{\uparrow}^{(1)}(0)$ ,  $N_{\downarrow}^{(1)}(0)$  - densities of the electronic states at the Fermi level in the ferromagnet for separate spin subzone. Then  $(C_{\uparrow} + C_{\downarrow}) = e^{-1} G^{FN}$ .

Spin polarization degree  $P$  of the ferromagnet is equal to:<sup>17</sup>  $P = \frac{N_{\uparrow}(0) - N_{\downarrow}(0)}{N_{\uparrow}(0) + N_{\downarrow}(0)} = \frac{C_{\uparrow} - C_{\downarrow}}{C_{\uparrow} + C_{\downarrow}}$ .

For small bias applied to the junction there is a nonequilibrium quasiparticles distribution function in superconductor  $f(E_k)$  which can be described by the equilibrium Fermi-function  $f_0(E_k)$  with the nonequilibrium additive term  $\pm \delta\mu_F$  to chemical potential for two spin subsystems:  $\mu_{\uparrow} = \mu + \delta\mu_F$ ,  $\mu_{\downarrow} = \mu - \delta\mu_F$ .<sup>18</sup> For  $eV \ll \Delta$  the value of this term is much smaller than the energy gap  $\Delta$  and linearly depends on the bias.<sup>19</sup> Thus the charge and the spin currents in the junction are as follows:

$$I_{eV \ll \Delta}^{FS} = (C_{\uparrow}(eV - \delta\mu_F) + C_{\downarrow}(eV + \delta\mu_F))\sigma^{NS}(T) = (1 - P\kappa)\sigma^{NS}(T)eVG^{FN} \quad (4)$$

$$I_s^{FS}|_{eV \ll \Delta} = (C_{\uparrow}(eV - \delta\mu_F) - C_{\downarrow}(eV + \delta\mu_F))\sigma^{NS}(T) = (P - \kappa)\sigma^{NS}(T)eVG^{FN} \quad (5)$$

where  $\kappa = \frac{\delta\mu_F}{eV}|_{eV \ll \Delta}$ .

Let us designate  $\alpha \equiv \frac{I_s^{FS}}{I_{eV \ll \Delta}^{FS}} = \frac{P - \kappa}{1 - P\kappa}$ . Then, taking into account (2)-(5), the normalized conductivity of F-I-S junction  $\sigma^{FS}$  can be calculated as follows:

$$\sigma^{FS}(P, T) = \frac{1}{G^{FN}} \frac{I_{eV \ll \Delta}^{FS}}{V}|_{eV \ll \Delta} = (1 - P\kappa)\sigma^{NS}(T) = \frac{(1 - P^2)}{1 - \alpha P^2} \sigma^{NS}(T) \quad (6)$$

From dependence (6) we see that the value of the normalized tunnel F-I-S junction conductivity  $\sigma^{FS}$  depends on the ferromagnet spin polarization degree  $P$  and the value  $\kappa$  in superconductor (or ratio  $\alpha$  of the charge current value  $I_{eV \ll \Delta}^{FS}$  and the spin current value  $I_s^{FS}$ ). So the value of  $\sigma^{FS}$  can differ essentially from the N-I-S tunnel junction normalized conductivity  $\sigma^{NS}$  calculated according to BCS theory or measured in experiments.

It is known that the presence of excess quasiparticles in N-I-N tunnel junctions results in insignificant (about few percent) increase in differential resistance at zero bias.<sup>20</sup> The observed effect is connected with the excess quasiparticles occupying the initially free energy states, which reduces tunneling probability for electrons. The quantity of excess quasiparticles is connected with time of their relaxation on low-energy phonons, whose density is insignificant.

Aronov<sup>21</sup> has theoretically shown that the tunneling current in ferromagnet-superconductor junction leads to spin polarization of quasiparticles in a superconductor. Both the spin polarization of quasiparticles in the superconductor and the external injection of spin polarized current leads to accumulation of the excess nonequilibrium spin polarized quasiparticles. The physical reason of this phenomenon is connected with the fact that spin

polarized electrons can not directly recombine into singlet Cooper pairs. For them to recombine, the electron spin flip processes are preliminary required. The probability of such processes in the superconductor in the absence of the magnetic impurities is extremely small.<sup>21</sup> Presence of the excess quasiparticles in the superconductor in addition to the thermal ones can result in lower conductance of the tunnel junction.

In our investigated F-I-S Co<sub>2</sub>CrAl-I-Pb film structures in the superconducting Pb there are no possibilities for effective flip-processes of the injected spin-polarized quasiparticles. That is why there is a possibility of accumulation of nonequilibrium spin-polarized electrons and their occupying the initially free energy levels in the superconductor, and, as a result, blocking of tunneling process from the ferromagnet. This will reduce the conductivity of the tunnel contact  $\sigma^{FS}$ .

Reduction of  $\sigma^{FS}$  in comparison with  $\sigma^{NS}$  due to the spin blocking of the tunneling process will take place only if the effective life-time for the spin-flip processes  $\tau_{sf}$  for the spin-polarized electrons in the junction region is longer than the electron life-time at a given temperature for the tunneling through the barrier  $\tau_T \sim \frac{1}{v_{k\perp} P(v_{k\perp})}$ , where  $P(v_{k\perp})$  is probability of the electron transfer through a barrier,  $v_{k\perp}$  - component of the electron's velocity normal to the barrier.<sup>14</sup>

The ratio of the number of electrons which undergo the spin-flip process to the total number of electrons, which pass through the barrier into the superconductor equals  $\tau_T/\tau_{sf}$ . Spin-flip doubles the portion of electrons capable to recombine. It will increase the number of free energy levels and will reduce spin polarization degree of excess quasiparticles in a superconductor. Magnitude of spin depolarization effect due to this mechanism can be characterized by the *factor of recombination of spin depolarization*  $\Theta = 2\tau_T/\tau_{sf}$ . Parameter  $\Theta$  defines the portion of electrons which recombine into singlet Cooper pairs due to the spin-flip as a part of difference between the number of electrons which have tunneled into a superconductor with major and minor spin projections.

Multiplier  $(P - \kappa)$  in (5) gives the decrease of  $I_s^{FS}|_{eV \ll \Delta}$  with increase of  $\kappa$ . On the other hand, this value is equal to  $\Theta P$ . Equating the two expressions, one can obtain:  $\kappa = P(1 - \Theta)$ ,  $\alpha = \frac{P - \kappa}{1 - P\kappa} = \frac{P\Theta}{1 - P^2(1 - \Theta)}$  and

$$\sigma^{FS}(p, T) = (1 - p^2(1 - \Theta))\sigma^{NS}(T) \quad (7)$$

In Fig.4 it is shown a set of lines of possible  $(\sigma^{FS}/\sigma^{NS})^{-1}$  values in F-I-S junction depending on  $P$  at different values of  $\Theta$ .

In the case with zero spin current ( $\alpha = 0$ ):<sup>16</sup>  $\sigma^{FS} = \sigma^{NS}(1 - P^2)$ . If in this case  $P = 1$ , then  $\sigma^{FS} = \sigma^{NS}(1 - P^2) = 0$ , i.e. injection current is absolutely blocked.

The obtained theoretical results can be used for experimental determination of spin polarization degree in ferromagnets.

For two different F-I-S junctions with the same ferromagnet dependence (7) gives  $\sigma_{1,2}^{FS}(p, T) = \sigma^{NS}(T)(1 -$

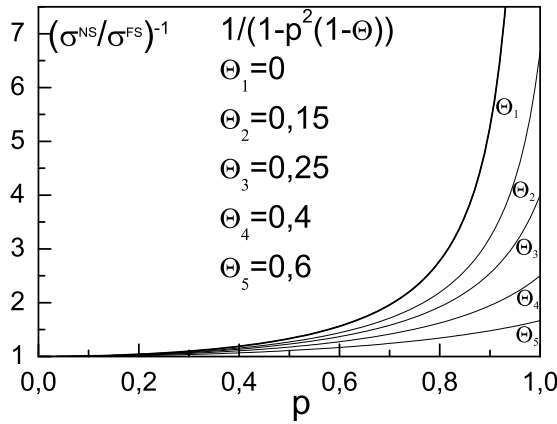


FIG. 4: Dependence of the normalized conductivity of F-I-S junction  $\sigma^{\text{FS}}$  on the spin polarization degree  $P$  for different values of the recombination of spin depolarization parameter  $\Theta$  and for different values of  $\alpha$ .

$p^2(1 - \Theta_{1,2})$  (we assume that  $\sigma_1^{\text{FS}} < \sigma_2^{\text{FS}}$  and, correspondingly,  $\Theta_1 < \Theta_2$ ). Having determined the values of the normalized conductivities  $\sigma_{1,2}^{\text{FS}}$  of these junctions from the experiment, one can determine spin polarization degree of this ferromagnet as follows:

$$1 - P^2 = \frac{\sigma_1^{\text{FS}}}{\sigma^{\text{NS}}(T)} \frac{\frac{\Theta_2}{\Theta_1} - \frac{\sigma_2^{\text{FS}}}{\sigma_1^{\text{FS}}}}{\frac{\Theta_2}{\Theta_1} - 1} = \frac{\sigma_1^{\text{FS}}}{\sigma^{\text{NS}}(T)} \frac{\frac{G_1^{\text{FN}}}{G_2^{\text{FN}}} - \frac{\sigma_2^{\text{FS}}}{\sigma_1^{\text{FS}}}}{\frac{G_1^{\text{FN}}}{G_2^{\text{FN}}} - 1}, \quad (8)$$

where accounted  $\frac{\Theta_2}{\Theta_1} = \frac{\tau_{T2}}{\tau_{T1}} = \frac{P_1(v_{k\perp})}{P_2(v_{k\perp})} = \frac{G_1^{\text{FN}}}{G_2^{\text{FN}}}$ .<sup>14</sup>

We have measured the values of  $G^{\text{FN}}$  and  $\sigma^{\text{FS}}$  for different  $\text{Co}_2\text{CrAl}$ -I-Pb junctions formed with the films of the same half-metal ferromagnet  $\text{Co}_2\text{CrAl}$  (Fig.1). Using different pairs of values  $G_i^{\text{FN}}$  and  $\sigma_i^{\text{FS}}$ , we have determined

the spin polarization degree  $P$  of this half-metal ferromagnet  $\text{Co}_2\text{CrAl}$  and obtained  $P = 0.97 \pm 0.03$ . In (8), we used calculated from the BCS theory value  $(\sigma^{\text{NS}})^{-1} = 6.5$  for temperature  $T = 4.2$  K. Theoretical curve (7) for determined  $P = 0.97$  and  $\Theta \sim (2G^{\text{FN}})^{-1}$  is shown in Fig.3 and is in good agreement with the experimental data.

As we see, the value of spin polarization degree of the ferromagnetic half-metal  $\text{Co}_2\text{CrAl}$  is  $P = 0.97 \pm 0.03$ , that only slightly differs from the theoretical value  $P_t = 1$ .

#### IV. CONCLUSIONS

1. The phenomenon of spin blocking of the tunnel current in  $\text{Co}_2\text{CrAl}$ -I-Pb junctions, which leads to a change in the normalized differential conductivity of the junctions at zero bias is observed. The value of spin blocking depends on the ferromagnet spin polarization degree  $P$  and the value of the factor of recombination of spin depolarization  $\Theta = 2\tau_T/\tau_{\text{sf}}$ .

2. It is established that the normalized conductivity  $\sigma^{\text{FS}}$  of  $\text{Co}_2\text{CrAl}$ -I-Pb tunnel junctions can be essentially smaller than the normalized conductivity  $\sigma^{\text{NS}} = 0.15$  of N-I-S tunnel junction. The value of  $\sigma^{\text{FS}}$  depends on the ferromagnet spin polarization degree  $P$  and the value of junction conductivity in the normal state  $G^{\text{FN}}$ . It is revealed that  $\sigma^{\text{FS}}$  can be smaller than 0.01 for tunnel junctions made of quasimonocrystal films of ferromagnetic half-metal Heusler alloy  $\text{Co}_2\text{CrAl}$ .

3. We have fabricated quasimonocrystal films of ferromagnetic half-metal Heusler alloy  $\text{Co}_2\text{CrAl}$  with the spin polarization degree  $P = 0.97 \pm 0.03$ , that is close to the theoretical value  $P_t = 1$ .

4. It is shown that measuring the differential conductivity in tunnel junctions of ferromagnet-insulator-superconductor type at zero bias allows us to determine the ferromagnet spin polarization degree.

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